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Photoacoustic Observation of Thermal Wave Interference Effects in Thin Samples

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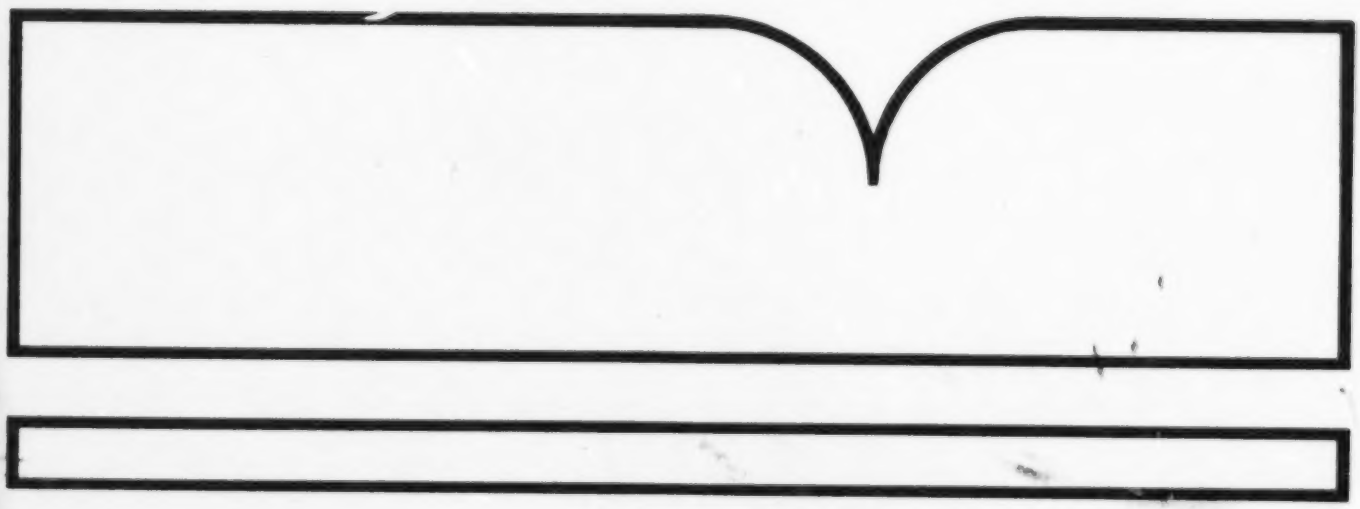

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16. ABSTRACT

The photoacoustic response arising from very thin and highly adsorbing layers contains structure resulting from thermal wave interference effects within the samples. The nature of these effects is dependent upon the chopping frequency and upon the thermal properties of the backing material. This structure provides information that may prove to be useful in analyzing the thickness or thermal properties of such layers. As applied to detection of soot collected from this ambient air, the thermal wave interference phenomenon is one factor in relating photoacoustic response versus soot loading on a filter.

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Photoacoustic observation of thermal wave interference effects in thin samples

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During the course of a photoacoustic study of ambient elemental carbon, this laboratory had the opportunity to prepare a set of accurately weighed and evenly deposited samples of laboratory-generated elemental carbon particles on Teflon filter substrates; we observed a dependence of photoacoustic signal on chopping frequency that must be recognized in analyzing ambient filters and that may prove to be a valuable tool for studying thicknesses and thermal properties of thin film samples. In this Communication, we relate this frequency-dependent structure in the photoacoustic signal to interference effects of the thermal waves within the very thin carbon samples. The formula of Rosencwaig and Gersho[1] is modified to illustrate the way in which the thermal properties of the backing material influence these interference effects. A thorough understanding of these phenomena is essential if photoacoustic data obtained from very thin and highly absorbing samples are to be properly interpreted.

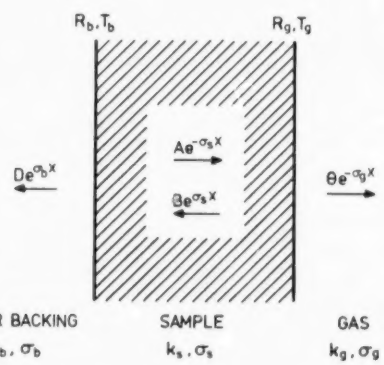

Fig. 1. Thermal waves in each of the three regions of the photoacoustic cell. Light is incident from the right.

Since elemental carbon is so highly absorbing (absorption coefficient $\beta > 10^5$ cm^{-1}), the theory of Rosencwaig and Gersho[1] is appropriate for the description of the photoacoustic response. The photoacoustic signal arises from the energy absorbed from an amplitude modulated beam of light that impinges upon the sample. This absorbed energy initiates rapidly damped traveling thermal waves[1] within the sample, and the resulting thermal wave that is transmitted to the gas within the photoacoustic cell produces the acoustic signal. If the sample is thin enough, thermal wave interference will affect the amplitude of this transmitted thermal wave and hence will affect the photoacoustic response.[2,3]

The thermal waves in the filter backing, sample, and gas regions of the photoacoustic cell are illustrated in Fig. 1. In each region $\sigma_i = (1 + j)a_i$, where $j = \sqrt{-1}$, and where a_i is the wave number of the thermal wave in the region i. As well as being dependent upon the thermal properties of the material in which the thermal wave is propagating, a_i is dependent upon the angular frequency ω at which the incident light beam is modulated; in particular, $a_i \propto \sqrt{\omega}$ or equivalently, $\lambda_i \propto 1/\sqrt{\omega}$ where λ_i is the wavelength of the thermal wave in the region i. Therefore, we may vary the wavelength of the thermal waves in each of the three regions by varying the rate at which the incident light beam is modulated. The thermal waves initiated within the sample travel to the sample-backing and sample-gas boundaries. By applying the boundary conditions of temperature and flux continuity we may compute the thermal wave reflection coefficient R_i and the thermal wave transmission coefficient T_i at each of these boundaries. These coefficients have the form[2]

$$R_b = \frac{1-b}{1+b}\,,\quad T_g = \frac{2}{1+b}\,,\quad R_g = \frac{1-g}{1+g}\,,\quad T_g = \frac{2}{1+g}\,,$$

where

$$b = \frac{k_b\sigma_b}{k_s\sigma_s}\,,\quad g = \frac{k_g\sigma_g}{k_s\sigma_s}\,,$$

and where k_i is the thermal conductivity in (W/cm K) in the medium i and σ_i is defined above. It is important to note that thermal waves are rapidly damped; in one wavelength the wave will be attenuated by a factor of $e^{-2\pi}$. The backing and gas regions of the cell are assumed to be thicker than one thermal wavelength in that region so that there is only a transmitted right traveling wave in the gas and a transmitted left traveling wave in the backing. However, if the sample is less than a thermal wavelength in thickness, the thermal waves within the sample will not be completely attenuated before multiply reflecting between the two boundaries, resulting in thermal wave interference that will affect the amplitude of the transmitted wave.

Apart from some minor algebraic modifications that illustrate the present discussion, the amplitude of the thermal wave transmitted to the gas as derived by Rosencwaig and Gersho[1] is

$$\theta = \frac{I_0\beta T_g}{4k_s(\beta^2 - \sigma_s^2)}\left\{\frac{(r-1)\{1-\exp[-(\beta+\sigma_s)l]\} + (r+1)R_b\exp(-2\sigma_s l)\{1-\exp[-(\beta-\sigma_s)l]\}}{1 - R_bR_g\exp(-2\sigma_s l)}\right\},$$

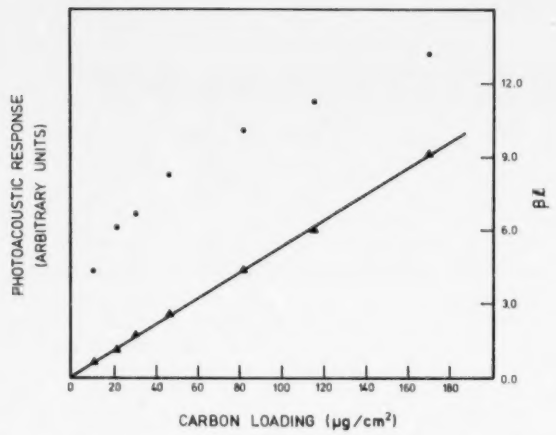

Fig. 2. Photoacoustic signal and optical thickness βl plotted vs carbon loading for a 100-Hz chopping frequency. ● Photoacoustic signal; ▲ optical thickness.

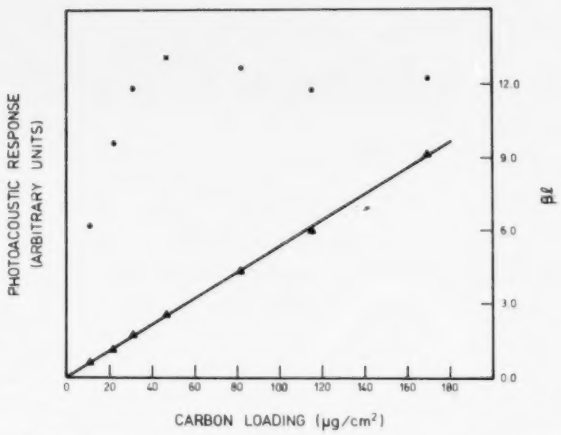

Fig. 3. Photoacoustic signal and optical thickness βl plotted vs carbon loading for a 3000-Hz chopping frequency. ● Photoacoustic signal; ▲ optical thickness.

where I_0 = incident light intensity in (W/cm^2),
β = absorption coefficient in (cm^{-1}),
l = thickness of the sample in (cm),
$r = \beta/\sigma_s$,
and where k_s, σ_s, T_g, R_g, and R_b are defined above. The first term in the large braces represents the contribution from the thermal waves that initially travel towards the gas, and the second term represents the contribution from the thermal waves that initially travel towards the backing; hence the factor R_b. Whether or not there is a 180° phase shift for thermal waves reflecting from the sample-backing boundary depends upon the sign of R_b[2,3]; this will be an important consideration in interpreting our photoacoustic results.

The detector used in these measurements consisted of a small cyclindrical cell 1.3 cm in diameter and 0.3 cm long equipped with a Knowles model BT 1759 microphone. The entrance window was glass and the output window was opal glass. An RCA 1P39 photodiode, placed immediately behind the opal glass, was used to measure the fraction of light transmitted by each filter. White light collected from a tungsten filament source was modulated with a PAR model 192 variable speed chopper prior to impinging on the carbon particles contained on Teflon filter substrates mounted in the photoacoustic cell. The output of the microphone was measured by a PAR model HR8 lock-in amplifier, and the output of the photodiode was measured with a digitial voltmeter.

Figure 2 shows a plot of photoacoustic response vs carbon loading obtained at a 100-Hz chopping frequency. Also plotted in Fig. 2 is the optical thickness βl, obtained from the transmission measurements, vs mass loading. Note that the photoacoustic signal does not saturate despite the fact that the last three filters in Fig. 2 are highly opaque; this indicates that the thermal waves reflecting from the sample-backing boundary are shifted in phase by 180° so that the intensity of the wave transmitted to the gas continues to rise as the sample thickness approaches (but, for the 100-Hz chopping frequency, has not reached) a thickness for which constructive interference would exist.

The photoacoustic response obtained at a 3000-Hz chopping frequency is plotted in Fig. 3. The same samples were used for both Figs. 2 and 3. Increasing the chopping frequency has decreased the wavelength of the thermal waves within the carbon samples so that more structure is revealed as the sample thickness increases. Again, the thermal waves at the sample-gas boundary interfere destructively at first, then constructively, then destructively, and so on. The last sample in Fig. 3 should be very close to saturation since it is approaching one thermal wavelength in thickness (unlike the optical case, the attenuation within the sample leads to a rapidly damped oscillation about the saturation level).

In conclusion, the photoacoustic response arising from very thin and highly absorbing layers contains structure resulting from thermal wave interference effects within the sample. The nature of these effects is dependent upon the chopping frequency and upon the thermal properties of the backing material. This structure provides information that may prove to be useful in analyzing the thickness or thermal properties of such layers.

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